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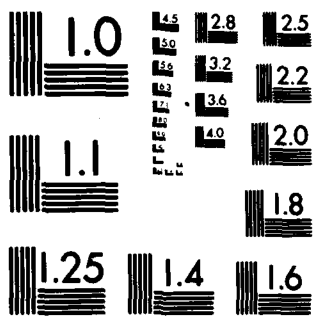

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FINAL REPORT
OF
MOLECULAR BEAM STUDIES OF LOW ENERGY REACTIONS

PERIOD: 1 SEPTEMBER 1973-31 JANUARY 1980

ONR CONTRACT NO. N00014-74-C-0011

PRINCIPAL INVESTIGATOR: R. H. NEYNABER

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IRT Corporation

Instrumentation
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7650 Convoy Court • P.O. Box 80817
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

A summary of research performed under ONR Contract No. N00014-74-C-0011 is given. The contract covered the period 1 Sept. 1973-31 Jan. 1980. The report describes merging and crossed-beams studies of chemi-ionization and/or ion-molecule reactions. Included are investigations of the Ne^*-Ar, Ne^*-Kr, Ne^*-Xe, He^*-D, Ar^*-Na, He^*-He^*, He^*-Ne^*, Ar^*-Kr^*, and He^+-Ne^* systems.

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Final Report

of

Molecular Beam Studies of Low Energy Reactions

ONR Contract No. N00014-74-C-0011

1. Contract Description

Chemi-ionization and ion-molecule reactions involving metastable and ground-state atoms are studied by both crossed and merging beams at low relative energies (i.e., 0.01 to 10 or 20 eV).

2. Scientific Problem

Some theories exist for chemi-ionization involving collisions of metastable and ground-state rare gases. There are very little experimental data to test these theories over a range of relative kinetic energy from 0.01 to 10 or 20 eV. We will supply such data. Theoretical work for collisions between two metastables is almost non-existent, and experimental data are scant. We will supply experimental information such as absolute and relative cross sections and branching ratios for associative to Penning ionization. This information should establish patterns to test those calculations that do exist and will stimulate further theory. Our chemi-ionization data also will produce some information on unknown potentials for the systems A^*B and C^*D^*, where A, B, C, and D are atoms and asterisks denote metastables. This information includes well depths and the dependence of the long range potential on internuclear separation.

The composition of keV neutral rare gas beams formed by charge transfer of the rare gas parent ion beam in alkalis is unknown. The beams consist of rare gas metastables (generally in two states) and ground-state atoms. The technique for generating such beams is common, and information on the composition is needed in analyzing data obtained through their use. We have developed a method for obtaining the fraction of ground-state atoms in such beams by studying appropriate ion-molecule reactions. We will apply this

method to determine unknown compositions.

No experimental information exists on low-energy resonant or near-resonant charge-transfer reactions between rare gas ions and metastables. Our experiments will supply such information. The data can be used to see if existing theories for charge transfer between ions and ground-state atoms can be extended to this case. We also will investigate energy distributions of product ions from which information on the reaction kinetics can be obtained.

3. Scientific and Technical Approach

Merging-beams techniques will be used for most of the studies. The two reactants of the process under investigation will be merged. Their velocities will be adjusted with respect to each other so that the desired relative energy in the center-of-mass system will be obtained. Product ions resulting from the reaction will be collected to give relative and absolute cross sections, and branching ratios will be obtained when appropriate.

Some crossed-beams measurements will be made of the ion-molecule reactions at relative energies above 1 eV. Again product ions will be measured to obtain cross sections.

4. Results

4.1 Chemi-ionization Studies

Our chemi-ionization studies have been largely of Penning ionization (PI) and associative ionization (AI) using merging-beams techniques. Most of the effort has been devoted to processes in which both reactants are atoms. Some of these reactions can be described as

$$R^* + A \rightarrow RA^+ + e \quad \text{(AI)} \qquad (1)$$

$$\rightarrow R + A^+ + e \quad \text{(PI)} \quad , \qquad (2)$$

where R^* is a metastable rare gas and A is a ground-state atom. These reactions are heteronuclear, and, hence, R is different from A. Although we have conducted some studies in which A was N or O, we will concentrate the discussion on those cases where A is a rare gas, atomic deuterium, or Na.

The remainder of the AI and PI reactions studied can be characterized as

$$R^* + R'^* \rightarrow RR'^+ + e \quad \text{(AI)} \qquad (3)$$

$$\rightarrow R + R'^+ + e \quad \text{(PI)} \qquad (4)$$

$$\rightarrow R^+ + R' + e \quad \text{(PI)} \quad , \qquad (5)$$

where R'^* is a metastable rare gas, and R' can be the same as or different from R.

The $R^*(R'^*)$ represents a composite of the two metastable states of the rare gas. For He the metastable states are 2^3S and 2^1S at energies of 19.8 and 20.6 eV, respectively. For Ne, Ar, and Kr the two metastable states are $^3P_2$ and $^3P_0$ at energies of 16.62 and 16.71 eV, 11.55 and 11.72 eV, and 9.92 and 10.56 eV, respectively.

Studies of Reactions (1)-(5) were generally made in the range $0.01 \leq W \leq 10$ eV (where W is the interaction energy, i.e., relative kinetic energy of the reactants) by measuring the product ionic currents. In addition, laboratory energy distributions of Penning ions were obtained. From these measurements the following quantities were determined: absolute and relative cross sections for AI and PI (Q_{AI} and Q_{PI}), total ionization cross sections $Q_T(\equiv Q_{AI} + Q_{PI})$, $r \equiv Q_{AI}/Q_{PI}$, and the branching ratio $R(\equiv Q_{AI}/Q_T)$. In addition, information was obtained on the dynamics of the collision process and on the long-range form and the well depth ε^* of the potential curve $V^*(R)$ of the reactants.

An effort has been made to observe patterns in Q_{AI}, Q_{PI}, Q_T, and R that can be related to the attractive or repulsive nature of the reactant system. (A system is defined as attractive if $W/\varepsilon^* \ll 1$, where W is in the thermal energy range, i.e., near 0.05 eV, and repulsive if $W/\varepsilon^* \gg 1$.) Such patterns could lead to some degree of predictability for the unmeasured Q and R of a given attractive or repulsive system.

The ideal situation would be to have a theory that could be applied to obtain dependable Q's and R's. A few theoretical models have been devised and have been applied to some systems. In their more sophisticated forms these have been difficult to use even for simple systems and, in general, require a priori knowledge of $V^*(R)$, the product potential curve $V^+(R)$, and the coupling width $\Gamma(R)$ for $V^*(R)$ and $V^+(R)$. These functions are difficult to obtain. When comparisons of Q and R can be made with experimental results over a wide energy range (e.g., $0.01 \leq W \leq 10$ eV), they are often disappointing. Because of these

theoretical difficulties, There is some merit in our empirical approach to estimate, at least crudely, the Q and R of a given system.

Our chemi-ionization results can best be summarized in tabular form. Shown below are Tables I and II[1]. (References in these tables are given in Ref. 1 and reproduced in this report after the presentation of the tables.) Our rare gas beams not only consisted of metastables but also ground-state species. The measured fraction of ground-states species, f_g, is given in Table I. Theoretical fractions calculated using a statistical approach are designated as f_{gt}. The ground-state species do not contribute to chemi-ionization.

TABLE I. Fraction f_g of ground-state species in rare-gas beams formed by charge transfer of rare-gas ions at energy E in alkali vapor at pressure p.[a]

Rare gas[b]	Alkali	E (keV)	p^c (millitorr)	Range of p for const f_g (millitorr)[d]	f_{gm}[e]	f_g[e]	f_{gt}[f]
He	Cs	1.1	0.5	0.13-1.1	0.12	0.14	0.17
		1.6	0.6	0.5 -1.8	0.14	0.16	0.13
		4.0	0.5	0.06-2.0	0.12	0.14	0.13
He	Na	1.1	0.8	---	0.05	0.06	<0.02
		1.6	0.8	---	0.05	0.06	<0.02
		4.0	0.8	0.2-1.5	0.05	0.06	0.03
^{3}He	Na	5.8	0.8	---	0.11	0.13	0.10
Ne	Na	1.3[g]	0.48[g]	---	0.45	0.49 ± 0.04	0.42
		2.75	0.65	0.04-3.7	0.53	0.57 ± 0.05	0.50
		5.5	0.62	0.23-1.5	0.53	0.57 ± 0.05	0.50
Ar	Rb	1.0	0.15	---	0.56	0.61 ± 0.05	0.54
		2.0	0.15	---	0.58	0.62 ± 0.05	0.53
		4.0	0.6	---	0.62	0.66 ± 0.05	0.51
		5.0	0.6	0.12-1.0	0.60	0.63 ± 0.05	0.50
Kr	Cs	4.5	0.04	0.04-0.14	0.60	0.64 ± 0.07	0.60
		5.5	0.14	0.04-0.44	0.65	0.68 ± 0.06	0.56

[a] This table gives our most up-to-date values for f_g.

[b] Except when noted He denotes ^{4}He.

[c] Values are accurate to within a factor of 2. Length of charge-transfer cell is 10 cm.

[d] Presumably f_g is constant for p less than minimum of range. Measurements were not made where a range is not specified except as noted by footnote g.

[e] The γ^*/γ = 1 for f_{gm} and 1.16 for f_g. The γ's are defined in Ref. 10. The f_{gm} and f_g for He are upper bounds, see Ref. 10 for the explanation.

[f] The f_{gt} contain undesignated errors due to the use of calculated charge-transfer Q's.

[g] The p and E for this measurement result in multiple collisions and quenching of Ne^* in the cell. Thus, the associated f_{gm} and f_g are unduly large.

TABLE II. Data for various molecular systems

System[a]	Researchers[b]	s[c]	ϵ^*(eV)	W(eV) or equivalent[d]	P	$Q_I(10^{-16}cm^2)$[e] or $k(10^{-11}cm^3/sec)$[e]	Ref.
Ne^*-Ar	Neynaber & Magnuson	0.31±0.03	--	0.033	0.28±0.08	$30.5^{+12.6}_{-10}$	8(a)
	Tang et al	0.31±0.007	--	0.033	--	9.1±0.4[h]	14(a),14(b)
	Kramer et al	--	--	0.033	0.38±0.005	--	15
	West et al	--	--	435K	0.31±0.03	14.7±2.9	16
$Ne(^3P_2)$-Ar	Brom et al	--	--	300K	--	10	17
Ne^*-Kr	Neynaber & Magnuson	0.38±0.04	--	0.05	0.30±0.05	$12.6^{+6.2}_{-5.3}$	18
	Tang et al	0.36±0.007	--	0.05	--	8.20±0.4[h]	14(a),14(b)
	Kramer et al	--	--	330K	0.32±0.005	--	15
	Niehaus	--	--	0.05	0.32±0.02	--	19
	West et al	--	--	435K	0.30±0.04	17.6±3.5	16
$Ne(^3P_2)$-Kr	Brom et al	--	--	300K	--	11	17
Ne^*-Xe	Neynaber & Tang	0.46±0.05	--	0.05	0.24±0.04	14.0±5.2	13
				300K		k=9.8±3.6	13
	Tang et al	0.44±0.006	--	0.05	--	8.9±0.4[h]	14(a),14(b)
	Illenberger	--	--	0.05	0.20±0.02	--	20
	West et al	--	--	435K	0.24±0.03	14.0±5.2	16
$Ne(^3P_2)$-Xe	Brom et al	--	--	300K	--	12	17
		--	--	300K	--	k=7.4	17
$Ne(^3P_2)$-Xe	Huestis et al	--	--	300K	--	k=7.5±1	21
He^*-D	Neynaber & Tang	0.34	2.1	0.05	0.23±0.04	24.0±8	8(b)
$He(2^3S)$-H	Morgner & Niehaus	--	--	0.045	0.196±0.006	33.6±6.7	22
$He(2^3S)$-H	Fort et al	--	--	0.05	0.27±0.05	24.0±4	8(d)
$He(2^3S)$-D	Hickman & Morgner (Th)	--	--	0.05	0.16	36.5	8(c)
$He(2^3S)$-H	Hickman & Morgner (Th)	--	--	0.05	0.20	34.0	8(c)
	Hotop et al	--	2.44	--	--	--	23
	Miller & Schaefer (Th)	--	1.91	--	--	--	24
	Hickman et al (Th)	--	2.07	--	--	--	25
Ar^*-Hg	Neynaber & Magnuson	0.33	0.3-0.4	0.033	0.08±0.017	31.2±15.5	26
$^3He^*$-He^*	Neynaber et al	0.38	0.6	0.033	0.046±0.006	112±30	12
He^*-He^*	Garrison et al (Th)	--	0.56-0.61	0.033	0.74	92	7(h)
	Orlecke et al	--	--	0.033	0.70±0.12	84±17	27
	Myers & Cunningham	--	--	0.033	--	101±25	28
Ne^*-Ne^*	Neynaber & Tang	--	0.5	0.033	<0.04±0.003[f]	$>97^{+46f}_{-39}$	29
Ar^*-Kr^*	Neynaber & Tang	0.40	0.2-0.4	0.033	0.035±0.007	308±62	30

[a]Unless specified He^* is $^4He^*$

[b]The work is experimental unless (Th) for theoretical is specified. The list of researchers is not complete in all cases

[c]The s is defined by $Q_I \propto W^{-s}$. The max W for validity is 0.03 eV for Ne^*-Ar, Ne^*-Kr, and Ne^*-Xe, 0.04 eV for Ar^*-Hg, 0.1 eV for He^*-D and $^3He^*$-He^*, and 10 eV for Ar^*-Kr^*

[d]This is the energy associated with the values of P and Q_I in the table

[e]Unless specified the value shown is Q_I. Otherwise it is the rate coefficient k at 300K

[f]These limits apply because P_I of Ne^* was measured but not P_I of Ne^*

[g]A recent remeasurement at 10° of Q_I(W=0.033 eV) for the Ne^*-Ar system resulted in a value about 10% less than shown

[h]This value is larger than that in Reference 14(a) to account for a more refined value of v as given in Reference 14(b)

REFERENCES FOR TABLES I AND II

[From R. H. Neynaber, "Merging Beams Experiments with Excited Atoms," Electronic and Atomic Collisions, ed. by N. Oda and K. Takayanagi (North-Holland Publishing Co., Amsterdam, 1980), pp. 287-300.

8. See, for example, (a) R. H. Neynaber and G. D. Magnuson, Phys. Rev. A11, 865 (1975); (b) R. H. Neynaber and S. Y. Tang, J. Chem. Phys. 69, 4851 (1978); (c) A. P. Hickman and H. Morgner, J. Chem. Phys. 67, 5484 (1977); (d) J. Fort, J. J. Laucagne, A. Pesnelle, and G. Watel, Phys. Rev. A18, 2063 (1978).

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Some comments about the results in these tables are:

a) The theoretical f_{gt} in Table I are in quite good agreement with the measured values indicating the validity of predicting the populations of the rare-gas states by a statistical approach.

b) Our experimental values of ε^* in Table II are for attractive systems and are in good agreement with the ε^* of others when comparisons can be made. Those systems with no values of ε^* are repulsive systems.

c) Except for He^*-D, the R (as seen in Table II) for attractive systems are quite small (generally < 10%) and, at thermal energies, considerably lower than the R for the repulsive systems.

d) From Table II one notes that the Q_T for R^*-R'^* systems [see Reactions (3), (4), and (5)] are considerably greater than those for R^*-A [see Reactions (1) and (2)] systems.

More details, additional comments, and explanations of some of the conclusions above have been presented.[1] Furthermore, comparisons of relative Q_T versus W and R versus W for the repulsive and attractive systems in Table II have been given. One interesting observation from such comparisons is that the Q_T and R curves are nearly the same for the repulsive Ne^*-Ar, Ne^*-Kr, and Ne^*-Xe systems, whereas for attractive systems such similarities do not exist.

Chemi-ionization studies under the contract have also been done on a few systems not shown in Table II. These are the He^*-H_2,[2] He^*-N,[3] and He^*-O[3] systems.

4.2 Ion-Molecule Studies

In addition to the chemi-ionization work the merging-beams apparatus has been used on this contract to study the charge transfer between $He^+(1S)$ and Ne^* in the energy range $0.1 \leq W \leq 500$ eV.[4] The cross section monotonically increases with W. The threshold for the reaction is near 0.1 eV. A modified Demkov approach is used to calculate cross sections, which agree very roughly with the experimental values above $W \approx 3$ eV. At low W the agreement is poor.

A crossed-beams study was also done on reactions of C^+ and O^+ with CO_2. The energy range of C^+ was from 1 to 500 eV.

5. Publications

Publications credited to the contract are listed below.

a. R. H. Neynaber and G. D. Magnuson, "Associative and Rearrangement Ionization in Collisions of Metastable Helium with H_2," J. Chem. Phys. 61, 749 (1974).

b. R. H. Neynaber and G. D. Magnuson, "Product Energies for $N_2^+ + O_2 \rightarrow N_2 + O_2^+$," J. Chem. Phys. 61, 3490 (1974).

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i. R. H. Neynaber and G. D. Magnuson, "Chemi-ionization in Collisions of Metastable Argon with Sodium," J. Chem. Phys. 67, 430 (1977).

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p. R. H. Neynaber and S. Y. Tang, "Chemi-ionization in Collisions of Metastable Argon with Metastable Krypton," J. Chem. Phys. 71, 3608 (1979).

6. <u>Extenuating Circumstances</u>

None

7. There were no unspent funds remaining at the end of the contract.

8. No graduate students or postdoctoral personnel have been associated with the contract.

9. During the contract, R. H. Neynaber also received partial support from the General Physics Division of the Air Force Office of Scientific Research.

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1. For more details see R. H. Neynaber, "Merging-Beams Experiments with Excited Atoms," Electronic and Atomic Collisions, ed. by N. Oda and K. Takayanagi (North-Holland Publishing Co., Amsterdam, 1980) pp 287-300.

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